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Induced Radioactivity and Ozone Production in Air from National Synchrotron Light Source (NSLS) Operation

An Assessment for NESHAPS (National Emission Standards for Hazardous Air Pollutants)

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I. Introduction

Induced activity in air can be created by beam losses occurring during injection or routine operation. The induced activity is primarily created from high-energy photon or neutron interactions with nitrogen or oxygen atoms, creating short-lived positron emitters such as ^{11}C, ^{13}N, or ^{15}O. In high intensity and high-energy machines, these radionuclides can reach significant levels and require engineering or administrative controls to reduce personnel exposure and releases to the environment. Because of the low power of the NSLS beams, it will be shown that these radionuclides are insignificant in this facility. The potential for air activation will be considered for two locations:

1. Beam transported to the X-ray or VUV injection line beam dumps, and
2. Loss of stored beam in the X-ray ring.

The operations and enclosures described below are located on the first floor in Building 725.

II. Booster Operation

During tuning of the booster synchrotron and during booster studies, the electron beam at energies up to 1 GeV will be extracted from the ring and transported to the injection line beam stop. This beam stop is 20 inches of copper and provides ~10 attenuation lengths for high-energy bremsstrahlung, thereby absorbing most of the photons created by the stopping electrons. As a result, air activation produced by forward directed bremsstrahlung will be insignificant. Only activation produced by high-energy neutrons will be considered. Booster operation is conducted 200 – 300 hours per year.

We will assume a beam of 1 x 10^{10} e/s at 1 GeV. The power (P) in the stopping beam is

$$P = I\,V = 1 \times 10^{10}\ \text{e/s} \times 1\ \text{Amp}/6.25 \times 10^{18}\ \text{e/s} \times 1 \times 10^{9}\ \text{V} = 1.6\ \text{watts} \qquad (1)$$

The dose equivalent[1] for forward directed high-energy neutrons (HEN) (E> 100 MeV) component from electrons stopping in a thick iron target can be written as:

[1] **A Guide to Radiation and Radioactivity near High Energy Particle Accelerators**; A.H. Sullivan, Nuclear Technology Publishing 1992; p.80

$$H(HEN) = 4.6 \times 10^{2} \text{ Rem/hr/KW at 1 m} \quad (2)$$

Or

$$H(HEN) = 4.6 \times 10^{2} \text{ Rem/hr/KW} \times 1.6 \times 10^{-3} \text{ kW} = 736 \text{ mRem/hr} \quad (3)$$

This value[2] is reduced by 1/3 since the energy of this beam is 1 GeV and the value in equation 2 is for higher energies, or

$$H(HEN) = 1/3 \times 736 \text{ mRem/hr at 1 m} = 245 \text{ mRem/hr @ 1 m} \quad (4)$$

For neutron with E > 100 MeV, the fluence to dose equivalent conversion factor is:

$$5.6 \text{ n/cm}^2\text{/s} = 1 \text{ mRem/hr} \quad (5)$$

Therefore the HEN fluence Φ

$$\Phi(HEN) = 245 \text{ mRem/hr} \times 5.6 \text{ n/cm}^2\text{/s per mRem/hr} = 1.37 \times 10^{3} \text{ n/cm}^2\text{/s @ 1 m} \quad (6)$$

For simplicity of calculation, we conservatively assume that this fluence is isotropic rather than sharply peaked in the forward direction. We will calculate the activity created in a 1 m sphere of air around the interaction point.

The total volume V of air in a sphere of radius 1 m is:

$$V = 4/3\ \Pi\ r^3 = 4/3\ \Pi\ 1^3 = 4.2 \text{ m}^3 = 4.2 \times 10^{3} \text{ liters} \quad (7)$$

At STP, the number N of atoms of nitrogen and oxygen in that volume is:

$$6.02 \times 10^{23} \text{ molecules/mole} \div 22.4 \text{ l/mole} = 2.69 \times 10^{22} \text{ molecules/l} \times 4.2 \times 10^{3} \text{ l} \quad (8)$$

$$N = 1.13 \times 10^{26} \text{ molecules} \times 2 \text{ atoms/molecule} = 2.3 \times 10^{26} \text{ atoms} \quad (9)$$

in a sphere of air of 1 m radius.

We assume that the cross-section σ for ^{11}C and ^{13}N, is 10 mb and 40 mb for ^{15}O [3]. Therefore the total number N* of radioactive atoms of each species created per s is:

$$N^* = \Phi \cdot \sigma \cdot N = 1.37 \times 10^{3} \text{ n/cm}^2\text{/s} \times 10 \times 10^{-27} \text{ cm}^2 \times 2.3 \times 10^{26} \text{ atoms} = 3.15 \times 10^{3} \quad (10)$$

$$N^* = 3.15 \times 10^{3} \text{ atoms/s for } {}^{11}C \text{ and } {}^{13}N \quad (11)$$

[2] ibid; p.78, figure 3.2
[3] ibid, p.138 table 4.10

And

$$N^* = 1.26 \times 10^4 \text{ atoms/s for } {}^{15}O \quad (12)$$

The decay constant for each radionuclide is as follows:

Radionuclide	Half-life ($T_{1/2}$)	Decay constant (λ)
^{11}C	20 min	5.78×10^{-4} s^{-1}
^{13}N	10 min	1.16×10^{-3} s^{-1}
^{15}O	2.1 min	5.5×10^{-3} s^{-1}

The decay rate $dN^*/dt = -\lambda N^*$ and therefore, the production rates can be shown to be:

Radionuclide	Production Rate (pCi/s)
^{11}C	49.2 pCi/s
^{13}N	98.4 pCi/s
^{15}O	1870 pCi/s

At saturation, ^{11}C and ^{13}N will have an equal magnitude of 8.5×10^{-2} μCi; ^{15}O will total 3.4×10^{-1} μCi. Therefore, for operating times of an hour or longer, the total radioactivity T from the three positron emitters produced within one meter of the beam stop is:

$$T = 8.5 \times 10^{-2} \text{ μCi} \times 2 + 3.4 \times 10^{-1} \text{ μCi} = 5.1 \times 10^{-1} \text{ μCi} \quad (13)$$

Assuming uniform mixing of the radioactivity within the booster enclosure (volume ~ 500 m^3), the concentration C of the radioactive air within the room is:

$$C = 5.1 \times 10^{-1} \text{ μCi} \div 500 \times 10^6 \text{ cc} = 1.02 \times 10^{-9} \text{ μCi/cc of positron emitters} \quad (14)$$

The derived air concentration for immersion established by the DOE for occupational exposure is:

$$\text{DAC (positron emitters)} = 4 \times 10^{-6} \text{ μCi/cc}^{\,4} \quad (15)$$

The calculated saturation activity for the airborne emitters is 0.025% of the DAC. It is clear that no significant air activity will accumulate within the Booster synchrotron enclosure from this practice. It should also be noticed that this calculation is conservative in that higher beam currents, longer irradiation times and higher neutron fluxes are assumed than are realistic.

III. Storage Ring Operation

Operation of the storage ring is the second mode of NSLS operation that is evaluated for air activation. The X-ray storage ring operates at an energy of 2.8 GeV and stores up to 1.1×10^{12} electrons. Once the ring is filled, the beam will circulate with a typical lifetime of 18 – 20 hours,

[4] Part 835 Appendix C (1/1/99).

but normally the beam is dumped and refilled on a 12-hour cycle. Beam losses during stored beam and at beam dump are typically scattered at many locations around the ring. For this calculation, we will assume a refill every three hours and a loss at a single point.

The total power P of the beam averaged over 3 hours is:

$$P = IV = [1.1 \times 10^{12} \text{ e} \div (3 \text{ hours} \times 3.6 \times 10^{3} \text{ s/hr})] \times 2.8 \times 10^{9} \text{ V} \quad (16)$$

$$P = [1.02 \times 10^{8} \text{ e/s} \div 6.25 \times 10^{18} \text{ e/s/A}] \times 2.8 \times 10^{9} \text{ V} = 1.63 \times 10^{-11} \text{ A} \times 2.8 \times 10^{9} \text{ V} \quad (17)$$

$$P = 4.6 \times 10^{-2} \text{ watts} \quad (18)$$

It can be seen from the power level that this condition creates an average loss that is only ~ 3 % of that generated by the booster beam striking the injection stopper. It is clear from this comparison that the loss represented by the booster is a worse case.

IV. Environmental Dose/ Risk Assessment

As shown in above, the saturation concentrations within the booster enclosure are well below (0.025%) the DAC for occupational workers, and therefore it can be safely concluded that the dose to the members of the public would be negligible. However, an environmental risk assessment was necessary because any potential fugitive emissions could contribute to dose when emissions are outside of the accelerator enclosure. These fugitive emissions are evaluated to demonstrate compliance with the U.S. Environmental Protection Agency's (EPA) annual limit of 10 mrem to the members of the general public from DOE facilities operations. Again, the potential hazard to the general public from release of these small quantities of short-lived radionuclides is negligible; however, a dose/risk assessment was completed to demonstrate compliance with the EPA limits.

In normal stack emission evaluations the standard practice is to show compliance with NESHAPs regulations by characterizing the radioactivity released and then applying the dispersion models to estimate the effective dose equivalent to the members of the public. But because the emissions at NSLS are fugitive and there is no direct stack emission to the atmosphere, the activity released has to be estimated instead of being a direct stack measurement. There will also be fugitive emissions from the enclosures into the larger experimental building, and some subsequent mixing prior to environmental release from building ventilation, which will further lower the overall emission estimates. As shown in sections II and III above only very small quantities of short-lived radionuclides are produced within the enclosure for release to the environment. For the purpose of estimating the source term in the environment, a conservative assumption was made that the concentrations of the short-lived gases at the saturation production rates are the released activity into the environment during the NSLS operation. In other words, the concentration in the environment is the same as the Booster (an unlikely scenario).

Radioactive gases are produced by the interaction of bremsstrahlung photons/neutrons with the air nuclei. The airborne activity is in general short lived, and the total activation is much less than in solid shielding materials because the air mass exposed is much less than radiation lengths of solid required to stop the beam. Even without forced ventilation air exchange occurs and

therefore it not possible to accumulate more than a fraction of saturated activity of ^{13}N, ^{15}O, and ^{11}C radioactive gases produced. In estimating the source term released to the environment, the Booster is taken as the more limiting operations at the NSLS. The Booster is operated for about 200-300 hours (1.08E6 seconds) in a year and based on the previously calculated production rate of the short-lived gases; the source term for emissions was calculated.

Table 3. Source Term

Radionuclide	Concentration (Curies)
^{15}O	2 mCi
^{11}C	0.1 mCi
^{13}N	0.05 mCi

After estimating the source term, the dose calculations were performed using an EPA prescribed computer code. The CAP88-PC Model is based on the Gaussian plume model with its associated limitations.

The radiological dose and risk assessment to the maximally exposed individual (MEI; located downwind at the site boundary) was estimated using the Clean Air Act Code CAP88-PC, version 2.0 modeling program to show compliance with 40CFR 61.93 (a) regulations. The meteorological data (temperature, precipitation, wind speed, and mixing height) used in the modeling program were site specific. The agricultural assumptions were that 100 percent of vegetables were imported. Because Suffolk County does not have any dairy and cattle farms, 100 percent of milk and meat was imported from outside of the assessment area.

The total dose to the MEI resulting from the activation of air due to bremsstrahlung /neutron from the NSLS operation was estimated to be 1.12E-08 mrem/year. The potential dose was estimated to be well below the 0.1mrem/ yr. annual limit as specified in the 40 CFR 61, subpart H, which requires continuous monitoring under NESHAPs. Therefore, the dose/risk to the members of the public was minimal, and only an annual administrative review of the facility would be sufficient to evaluate any changes in operations, process, beam intensity, or any other factors that may increase emissions to the environment.

V. Ozone Generation

The full synchrotron radiation produced by the X-ray ring dipole bending magnets and certain insertion devices can generate significant ozone levels when the beam is allowed to pass through air. Traversal of the full beam through air can create ozone concentrations that may approach or exceed the ACGIH Threshold Limit Values for ozone. Controls are required to control potential exposures to workers and releases to the environment.

Transmission of the synchrotron beams from the storage ring to the experimental end stations occurs within either vacuum enclosures or enclosures containing an inert gas. These beam paths present no ozone concern. The experimental end stations in x-ray beam lines are enclosed in metal hutches that act as an exclusion zone for personnel. For some experiments, the synchrotron beam passes through air within these hutches and can produce ozone in these instances. The air path length, the energy spectrum and the flux of the beam, and the physical

size of the beam determine the amount of ozone generated. Several configurations have been implemented to reduce or eliminate the production of ozone inside these hutches. An outline of those controls follows.

- The entire experiment can be contained within an evacuated or inert gas-filled chamber, thus the full photon beam (commonly called a "white" beam) does not pass through air.
- The white beam path can also be contained within a "flight tube", i.e. a tube, sealed at both ends, under vacuum or filled with an inert gas, thus minimizing or eliminating exposure of the white beam to air.
- The beam dimensions can be minimized so that there is very little beam interacting with the air and the interactions are localized.
- If the experiment can tolerate such a change, the beam can be filtered to reduce the flux of the low X-ray energies that are responsible for the highest ozone production rates, i.e. use silicon to filter out the 3-15 keV X-rays, thus reducing the overall ozone production.
- The beam path length through air can be minimized and the air adjacent to the beam path can be scrubbed using an appropriate filter.

Ozone concentrations generated by an experiment are measured to determine potential exposures and a delayed entry time can be established when needed to allow the ozone concentration within the hutch to diminish to acceptable levels. In addition, where needed, hutches can be vented through charcoal filters to reduce concentrations within the hutch and to minimize releases into the experimental area.

These control features have worked well and ozone production and exposure have not proved to be a problem for workers entering the hutch or working in the experimental area around the hutch.

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Version 2.00

Clean Air Act Assessment Package - 1988

S Y N O P S I S R E P O R T

Non-Radon Population Assessment
Jan 12, 2004 02:16 pm

Facility: National Synchroton Light Source (NSLS)
Address: Building 725
P.O.Box 5000
City: Upton
State: NY Zip: 11973

Source Category: Area
Source Type: Area
Emission Year: 2004

Comments: NESHAPs Evaluation for NSLS

Effective Dose Equivalent (mrem/year)
1.12E-08

At This Location: 2000 Meters West Southwest

Dataset Name: NSLS
Dataset Date: Jan 12, 2004 02:16 pm
Wind File: C:\CAP88PC2\WNDFILES\BNL00.WND
Population File: C:\CAP88PC2\POPFILES\BNL98A.POP

NSLS REVISION & PERIODIC REVIEW LOG

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> See NSLS Quality Control Coordinator for original revision and review signatures <

REVISION TABLE

Rev	Description	Date
A	Original document	February 2004

PERIODIC REVIEW TABLE

Complete this table to record the completion of periodic reviews for an existing controlled document. A successful periodic review will reveal the existing document is current, correct, and does not require any revision/change.

Document Review Frequency
1 year

Rev	Date	Reviewed By (Print):	Signature:

Guidance from Balwan Hooda to N. Gmur, 4/28/2004:
An annual administrative review of the facility would entail a simple look at the NSLS operation to ascertain that process has not changed over the course of the year and as a consequence the dose to the members of the public.

This basically commits us to take another formal look (annually) at the systems/controls in place. It does not have to be a complicated committee level review but can be a simple straight forward statement that, "the Y process were reviewed and there are no deviations from the parameters of NESHAPs evaluation completed in CY XX." However, this example statement is with the assumption the beam was not upgraded or any other operational changes, etc. Also, it is an administrative review, therefore can be done by Bob Casey or yourself to document any observations during the review cycle.